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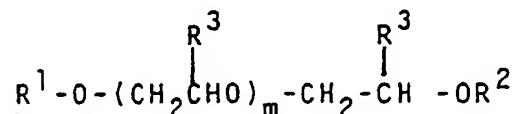
(54) Flowable graft and derivatized polymer concentrate and lubricant containing same.

(57) The bulk viscosity of an oil additive concentrate which comprises:

(a) a mineral oil of lubricating viscosity, and

(b) from 5 to 35 weight percent, based on (a) + (b), of a graft and derivatized polymer, can be reduced from a value above 2000 centistokes by incorporating a minor amount of a co-solvent having the formula:

a)



or

b) R<sup>4</sup>O-CO-CH<sub>2</sub>-(CH<sub>2</sub>)<sub>n</sub>-COOR<sup>4</sup>

wherein R<sup>1</sup> and R<sup>2</sup>, which can be the same or different, each represents hydrogen or a C<sub>1</sub> - C<sub>25</sub> hydrocarbon group,

R<sup>3</sup> represents hydrogen or a C<sub>1</sub> - C<sub>25</sub> hydrocarbon group,

each R<sup>4</sup> represents a C<sub>1</sub> - C<sub>12</sub> hydrocarbon group,

m is 0 to 25, and

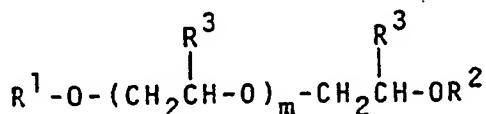
n is 0 to 10.

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or ether alcohols and ester of diacids of formulae (a) and (b) described hereinbelow.

The polymer substrates which are used in the preparation of the grafted and derivatized polymers comprising the oil concentrates employed in the instant invention will generally have a number average molecular weight from 5,000 to 500,000, or higher. More often, the polymer substrates intended for use in preparing multi-functional lubricating oil additives will have a number average molecular weight from 25,000 to 250,000. A particularly preferred class of polymer substrates are the ethylene-propylene copolymers having a number average molecular weight from 50,000 to 200,000, with a still more preferred range being from 75,000 to 150,000.

The classes of co-solvents which are employed to produce flowable oil concentrates of the high molecular weight grafted and derivatized polymers have the general formulae: a)



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in which R<sup>1</sup> and R<sup>2</sup>, which can be the same or different, each represent hydrogen or a hydrocarbon group having from 1 to 25, preferably from 1 to 12, carbon atoms and m is 0 to 25, preferably 0 to 10

20 and

b) R<sup>4</sup>O-CO-CH<sub>2</sub>-(CH<sub>2</sub>)<sub>n</sub>-COOR

in which each R<sup>4</sup> represents a hydrocarbon group having from 1 to 12 carbon atoms and n is 0 to 10.

In both formulae, the hydrocarbon groups R<sup>1</sup> to R<sup>4</sup> may be alkyl groups having from 1 to 5 carbon atoms, aryl groups having from 6 to 10 carbon atoms, or alkaryl groups having from 7 to 25 carbon atoms. The preferred values for m and n are 2 to 6.

25 Examples of the co-solvents represented by formula I above include the series of compounds designated as Surfonic Surface-active agents marketed by the Texaco Chemical Company. These compounds are prepared by reacting ethylene oxide with nonyl phenol to produce compounds represented by the formula: C<sub>9</sub>H<sub>19</sub>C<sub>6</sub>H<sub>4</sub>O(CH<sub>2</sub>CH<sub>2</sub>O)<sub>m</sub>, H in which m is from 1 to 10.

30 Reduction in the high bulk viscosities of the oil solution of the grafted and derivatized polymers is achieved by mixing a minor amount of the prescribed co-solvent with the copolymer. In general, an oil concentrate of the grafted and derivatized polymer containing from 5 to 25 weight percent of the grafted and derivatized polymer dissolved therein and containing from 0.1 to 5 weight percent of the co-solvent, based on the total weight of the concentrate will exhibit substantially improved flow or fluid characteristics for the concentrate mixture. A preferred amount of co-solvent in the concentrate mixture is from 0.5 to 3 weight percent, based on the total weight of the concentrate, with the most preferred concentration of the co-solvent being from 1.5 to 3 weight percent. In the method described above, a flowable concentrate is obtained by mixing an effective or suitable amount of the co-solvent into the oil concentrate of the final grafted and derivatized polymer or copolymer.

35 Alternatively, the flowable oil concentrate may be prepared by employing the co-solvent with the derivatizing compound before the reaction between the derivatizing compound and the grafted polymer. After the derivatization reaction has been completed, the result will be a substantially improved flowable oil concentrate of the grafted and derivatized polymer.

40 The following Examples illustrate the practice of this invention.

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#### EXAMPLE 1

50 47.0 grams of a solid maleic anhydride-grafted polymer (rubber) in which the polymer substrate comprised 58 mole percent of ethylene and 42 mole percent of propylene, having a number average molecular weight of about 135,000, on which had been grafted 1.1 weight percent of maleic anhydride was dissolved at 160°C with mechanical stirring under a nitrogen blanket in 507 grams of solvent neutral mineral oil having a viscosity at 100°C of 100 SUS (20.4 cSt). After the polymer had dissolved, mixing was continued for an additional hour at 160°C.

55 1.1 grams of neat N-phenyl-1,4-phenylenediamine was added to the oil solution of the polymer and reaction was effected over 4 hours at 160°C under nitrogen. The reaction mixture containing the derivatized graft polymer was then cooled to 100°C and filtered. The polymer content of the oil-polymer concentrate

mixture was 8.5 weight percent.

An attempt was made to measure the Kinematic viscosity at 100°C of the grafted and derivatized polymer-oil concentrate, but it was too viscous to measure. It was estimated to have a Kinematic viscosity at 100°C of greater than 5000 Centistokes.

5 A variety of co-solvents were added, at 160°C for minutes, to the above-prepared grafted and derivatized polymer-oil concentrate. The co-solvents were blended into the concentrates by mixing at 160°C for 30 minutes. The viscosities of the concentrate mixtures containing the co-solvent was determined and the results are set forth in Table I below.

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TABLE I

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Co-Solvent Effect on the Viscosity of the Oil-Polymer Mixture of Example I		
Co-Solvent	Amount, wt.% (1)	Kin Vis @ 100°C
None	---	TVTM (2)
Jeffox PPG 2000(a)	1.8	1572
Emolien 2986(b)	2.8	1530
Surfonic N-31.5(c)	1.8	1497
Surfonic N-40(d)	1.8	1471

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(1) Weight percent of co-solvent in the oil-polymer concentrate based on the total weight of the concentrate.

(2) Too viscous to measure (Kin Vis @ 100°C approx > 5000 CSt).

(a) (poly ethylene-propylene glycol).

(b) (2-ethylhexyl diester of azelaic acid).

(c) (ethoxylated nonyl phenol with 3.15 moles of ethylene oxide per mole of nonyl phenol).

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(d) (ethoxylated nonyl phenol with 4.0 moles of ethylene oxide per mole of nonyl phenol).

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The following Examples illustrate the preparation of the grafted and derivatized polymer wherein the co-solvent was employed during the derivation reaction.

#### EXAMPLE II

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47.2 grams of a solid maleic anhydride grafted ethylene-propylene copolymer having a number average molecular weight of about 135,000 and having 1.1 weight percent of maleic anhydride grafted thereon, was dissolved in 507 grams of the solvent neutral base oil used in Example I at 160°C with stirring, while being maintained under a nitrogen atmosphere. A mixture of 1.1 grams of N-phenylphenylenediamine and 10 grams of Surfonic N-31.5 was prepared at 70°C and charged to the reactor containing the oil solution of the grafted ethylene-propylene copolymer. The derivatization reaction was conducted at 160°C under a nitrogen atmosphere for 2 hours. The reaction mixture was cooled to 100°C and screen filtered through a 100 mesh (0.15 mm) screen. The bulk viscosity of the reaction mixture concentrate was 1495 Centistokes Kinematic Viscosity at 100°C, as compared to the reaction product without the addition os Surfonic N-31.5, which was too viscous to measure

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#### EXAMPLE II

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255 grams of a solid maleic anhydride-grafted ethylene propylene copolymer having a number average molecular weight of about 135,000 and having 1.1 weight percent of maleic anhydride grafted thereon was dissolved in 2745 grams of the solvent neutral based oil at 160°C with stirring while being maintained

under a nitrogen atmosphere. A mixture of 5.0 grams of N-phenylphenylenediamine and 50 grams of Surfonic N-40 was prepared at 70 °C, and charged to the reactor containing the oil solution of the grafted ethylene-propylene copolymer. The derivatization reaction was conducted at 160 °C under a nitrogen atmosphere for 2 hours. The reaction mixture was cooled at 100 °C and screen filtered through a 100 mesh (0.15 mm) screen. The bulk viscosity of the reaction mixture concentrate was 1229 Centistokes Kinematic Viscosity at 100 °C, as compared to the reaction product without the addition of Surfonic N-40, which was too viscous to measure.

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EXAMPLE IV

72.0 grams of a solid maleic anhydride graft ethylene-propylene copolymer having an number average molecular weight of about 80,000 and having 0.85 wt.% of maleic anhydride grafted thereon was dissolved in 528 grams of solvent neutral base oil at 160 °C with stirring while being maintained under a nitrogen atmosphere. A mixture of 1.5 grams of N-phenylphenylenediamine and 10 grams of Surfonic N-31.5 was prepared at 70 °C and charged to the reactor containing the oil solution of the grafted ethylene-propylene copolymer. The derivatization reaction was conducted at 160 °C under a nitrogen atmosphere for about 2 hours. The reaction mixture was cooled to 100 °C and filtered through a 100 mesh (0.15 mm) screen. The bulk viscosity of the reaction mixture concentrate was 1195 Centistokes Kinematic Viscosity at 100 °C. The bulk viscosity of this reaction product without the addition of Surfonic N-31.5 was 2048 Centistokes Kinematic Viscosity at 100 °C. This further illustrates the effectiveness of the co-solvent when used in the derivatization reaction.

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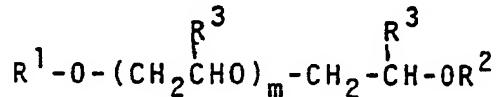
EXAMPLE V

The oil solution of the grafted ethylene-propylene copolymer of Example IV was reacted in a derivatization reaction with a mixture of 1.5 grams of N-phenyl-phenylenediamine and 10 grams of Surfonic N-40. The reaction was conducted in the same manner as in Example IV. The bulk viscosity of the reaction mixture concentrate was 1183 Centistokes Kinematic Viscosity at 100 °C.

35 **Claims**

1 . An oil additive concentratge which comprises:  
 (a) a mineral oil of lubricating viscosity, and  
 (b) from to 35 weight percent, based on (a) + (b) , of a graft and derivatized polymer,  
 40 characterised in that the bulk viscosity, measured as the Kinematic viscosity at 100 °C, of the concentrate is reduced from a value above 2000 centistokes by incorporating a minor amount of a co-solvent having the formula: a)

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or  
 b) R<sup>4</sup>O-CO-CH<sub>2</sub>-(CH<sub>2</sub>)<sub>n</sub>-COOR<sup>4</sup>  
 wherein R<sup>1</sup>and R<sup>2</sup>, which can be the same or different, each represents hydrogen or a C<sub>1</sub>-C<sub>25</sub> hydrocarbon group,  
 R<sup>3</sup> represents hydrogen or a C<sub>1</sub>-C<sub>25</sub> hydrocarbon group,  
 55 each R<sup>4</sup> represents a C<sub>1</sub>-C<sub>12</sub> hydrocarbon group,  
 m is 0 to 10, and  
 n is 0 to 10.  
 2. An additive concentrate according to Claim 1 characterised in that the amount of co-solvent is from

- 0.1 to 5 weight percent, based on the weight of concentrate.
3. An additive concentrate according to Claim 1 or 2 characterised in that the co-solvent has the formula:  
 $R^5\text{-Aryl-O-(CH}_2\text{CH}_2\text{-O)}_m\text{'H}$
- 5 in which  $R^5$  represents an aliphatic hydrocarbon radical and  $m'$  is 1 to 5.
4. An additive concentrate according to Claim 1 or 2 characterised in that the co-solvent has the formula:  
 $C_9\text{H}_{19}\text{C}_6\text{H}_4\text{O(CH}_2\text{CH}_2\text{O)}_m\text{'H}$
- in which  $m'$  is 1 to 10.
- 10 5. An oil concentrate according to any one of Claims 1 to 4 characterised in that the graft and derivatized polymer has a number average molecular weight of 5000 to 500,000.
6. An oil additive concentrate according to any one of Claims 1 to 5 characterised in that the graft and derivatized polymer is an ethylene copolymer grafted with maleic anhydride and derivatized with N-phenylphenylenediamine.
- 15 7. A lubricating oil composition comprising a mineral oil of lubricating viscosity characterised by a minor dispersant amount of the additive concentrate according to any one of Claims 1 to 6.

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## EUROPEAN SEARCH REPORT

Application Number

EP 89 31 1706

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 5)
Y	EP-A-0 190 869 (EXXON CHEMICAL PATENTS) * Page 5, line 17 - page 6, line 8; page 7, line 29 - page 8, line 7; page 9, line 16 - page 10, line 6; page 11, lines 1-7 * ---	1,2,5,7	C 10 M 161/00 C 10 M 157/00 C 10 M 157/04 // (C 10 M 157/00 C 10 M 145:26 C 10 M 145:36 )
Y	EP-A-0 014 746 (RÖHM GmbH) * Page 7, line 15 - page 8, line 28; page 13, example 1 * ---	1,2,5,7	(C 10 M 157/04 C 10 M 145:26 C 10 M 145:36 C 10 M 149:06 )
P,X	GB-A-2 206 600 (SANYO CHEMICAL INDUSTRIES) * Page 3, line 18 - page 4, line 2; page 4, lines 12-17; page 10, line 23 - page 14, line 7; page 16, lines 16-24 * ---	1,2,5,7	(C 10 M 161/00 C 10 M 129:08 C 10 M 129:16 C 10 M 129:72 C 10 M 149:06 ) C 10 N 30:02
D,A	EP-A-0 338 672 (TEXACO DEVELOPMENT CORP.) * Claims 1,7-10 * ---	6	
A	US-A-2 833 719 (W.L. VAN HORNE) * Claim 1 * ---	3,4	
P,A	EP-A-0 306 290 (EXXON CHEMICAL PATENTS) * Page 4, lines 26-47; claims 1,5,9,12,23 * -----	1,2,5,7	C 10 M
The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
THE HAGUE	06-02-1990	HILGENGA K.J.	
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone	T : theory or principle underlying the invention		
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